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Original Research

Morphological Pattern of Heavy Metal Zn in Soil and Its Driving Factors During Rice-Rapeseed Rotation

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Abstract

To study the speciation characteristics of soil Zn and its distribution in different tillage layers during paddy and drought rotation. Taking the cultivated soil under rice-rape rotation in Bozhou District, Zunyi City, Guizhou Province as the research object, the speciation characteristics and driving factors of soil Zn during rice-rape rotation were studied. In the process of rice-rape rotation, the total amount of heavy metal Zn in surface soil was greater than that in bottom soil. In the depth of 0-40 cm soil layer, the exchangeable fraction of Zn in soils was small, and residue fraction was the greatest. The exchangeable fraction of Zn in surface soils was positively correlated with carbonate bounded fraction and iron-manganese oxide bounded fraction, and negatively correlated with residue fraction. While the exchangeable fraction of Zn in bottom soil was positively correlated with iron-manganese oxide bounded fraction and organic bounded fraction, and positively correlated with residue fraction. During rice-rape rotation process, the content of Zn showed a significant surface aggregation, and was affected by different forms and contents.

Keywords: soil, heavy metal, rice-rape rotation, speciation, correlation

Introduction

In southern China, one of the common farming methods in agriculture is paddy-upland rotation, a planting method of rotating rice and upland crops successively on the same area of the soil [1, 2]. Rice-rapeseed rotation is a typical, paddy-upland crop rotation in certain areas. Due to factors such as water, climate, landform, and social development, most areas of Guizhou Province have adopted this farming method. Among major crops in China, rapeseed is an indispensable source of oil and energy. According to statistics from the Food and Agriculture Organization of the United Nations, the total production and planting area of rapeseed in China ranked first or second in the world for a long time. Rapeseed oil has always been a popular edible oil in daily life, and it accounts for more than 55% of various domestic oil crops [3]. Since the late 1990s, with the rapid social and economic development in the country, a large number of pollution problems

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have arisen. In recent years, factors that directly cause damage to the growth environment of crops, especially the pollution caused by heavy metals in the soil, have become increasingly evident, bringing potentially devastating impacts on agricultural development.

Zinc (Zn) is a widespread heavy metal that can be easily found on the surface of the earth, while the content of Zn was more than 100 mg·kg^{-1}, it can cause harm to organisms [4]. The heavy metal Zn is inevitably discharged into the surrounding soil and water in the industrial production process. In recent studies on soil and water, it has been found that heavy metal Zn can be transported and enriched in bio-organic tissues in large amounts along the food chain, posing a serious threat to the stability of the ecological environment of crops [5]. It will even be enriched in crops and passed along the food chain in biological organisms, eventually affecting human health [6]. There are many studies on heavy metal Zn, and most of them focus on the accumulation patterns of heavy metal Zn in soil under different rotation modes [7, 8], the evaluation of ecological restoration of heavy metal Zn-contaminated soil under different planting modes [9-11], or the source/sink patterns of heavy metal Zn under different planting modes [12]. However, there have been no studies involving the morphological characteristics of heavy metal Zn during paddy-upland rotation. Therefore, in this study, through field trial and analysis, the morphological pattern of heavy metal Zn in soil and its driving factors during the rice-rapeseed rotation were investigated, so as to fully explore the distribution pattern of heavy metal Zn under paddy and upland conditions and provide a theoretical basis for the treatment of resultant pollution.

Materials and Methods

General Background Information of the Researched Area

The trial site was located in Shiban Town, Bozhou District, Zunyi City, Guizhou Province, with an altitude of about 800m, longitude of 106°44'28" east, latitude of 27°30'11" north, annual average temperature of 14.9ºC, and annual precipitation of 1020.6 mm. With a subtropical monsoon humid climate, the area yields two crops a year in its calcareous rice soil. The rapeseed cultivar was Youyan No.57, which is a two-line hybrid cultivar of brassica oleracea semi-winter recessive genic sterility, and its national approval number is 2013001. The rice cultivar was Yixiang 725, which is a combination of Yixiang 1A and the self-selected restorer Mianhui 725 by the Mianyang Institute of Agricultural Science.

Design of Field Trial

The rice-rapeseed rotation was designed in accordance with local traditional farming methods. After rapeseed is harvested, the fields are filled with water to prepare for transplanting rice seedlings. The rapeseed is sown in mid-October, while the rice seedlings are cultivated in late April, planted in early June, and harvested in mid-September of the following year. The top soil (0-20 cm) and bottom soil (20-40 cm) surrounding the roots were sampled before rapeseed sowing and at its four growth periods (seedling development, Bolting, flowering, and harvesting periods), as well as before rice sowing and at its four growth periods (seedling, tillering, filling, and harvesting periods).

Sample Analysis

The morphology of heavy metal in soil was extracted following the Tessier sequential extraction procedure [13]. The total amount of heavy metal was extracted by the HF-HNO_3-$HClO_4$ digestion method. The content of heavy metal was determined by ICP-MS (2030, Shimadzu, Japan). In the experiment, the soil standard substance (GBW07403) and heavy metals were used to extract the morphological standard substance (GBW07436), in order to reduce the error and control the recovery rate. The recovery rates of all forms fell between 85% and 110%.

The activity of heavy metal in soil was expressed using the mobility factor (MF) of heavy metal by the equation as follows:

$$MF = \frac{EXC + CAR}{EXC + CAR + OX + OM + RES} \times 100\%$$

Where MF is the mobility factor; EXC is the exchangeable fraction; CAR is carbonate fraction; OX is iron-manganese oxide bound fraction; OM is organic-bound fraction; RES is residual fraction [14].

Results

Changes in the Total Heavy Metal Zn in Soil during Rice-Rapeseed Rotation

As shown in Table 2, the total heavy metal Zn in soil varied in different cultivated layers during the rice-rapeseed rotation. In the top soil, the total heavy metal Zn in soil varied insignificantly before sowing periods and at harvest periods of both rapeseed and rice. The total heavy metal Zn in soil fluctuated greatly at various growth periods of rapeseed, but the total amount at harvest period remained basically the same as before sowing. The total heavy metal Zn in soil fluctuated within the range of 75.22 mg·kg^{-1} to 76.33 mg·kg^{-1} at various growth periods of rice, and the change was insignificant. In the bottom soil, the total heavy metal Zn rose sharply and then slowly dropped at the seedling period of rapeseed, and finally the total amount at the harvest period remained basically the same as before

Table 1. Basic overview of the study area.

Soil Layers	Index	SOM (g·kg^{-1})	Eh (mV)	pH	Zn (mg·kg^{-1})
0-20 cm	Mean	56.19	10.20	7.80	81.72
	Standard deviation	2.38	12.62	0.09	1.23
	Coefficient of variation (%)	4.24	123.73	1.15	1.51
20-40 cm	Mean	41.03	29.20	8.07	69.49
	Standard deviation	2.64	6.72	0.10	0.57
	Coefficient of variation (%)	6.43	23.01	1.24	0.82

Table 2. total amount of heavy metal Zn in soil during rice-rape rotation (mg·kg^{-1}).

		Rape growth period				Rice growth period			
		Seedling stage	Bolting stage	Flowering stage	Harvesting periods	Seedling stage	Tillering stage	Filling period	Harvesting periods
0-20	Mean	73.98	79.99	70.94	82.16	75.22	76.33	74.01	75.87
	Standard deviation	1.06	1.14	0.76	0.65	0.99	1.30	1.27	1.25
20-40	Mean	78.21	74.18	71.36	70.17	69.64	73.55	71.08	78.29
	Standard deviation	0.80	1.58	0.41	1.48	0.99	1.36	1.52	1.95

sowing. The total heavy metal Zn increased in a staggered manner at the growth periods of rice, reaching a maximize of 78.29 mg·kg^{-1} at the harvest period.

Characteristics of Heavy Metal Contents in URM Producing Areas under Different Forms of Land Use

Changes in the Morphology of Heavy Metal Zn in Soil

As shown in Fig. 1, among the heavy metal Zn in the top soil during the rice-rapeseed rotation, the residual fraction accounted for the largest proportion (71.18%~80.10%), followed by the Fe/Mn oxide fraction (15.17%~22.21%), and the exchangeable fraction accounted for the smallest proportion (0.12%~0.31%). Compared with the growth period of rapeseed, the proportions of the exchangeable, carbonate-bound, and Fe/Mn oxide fractions all increased at the growth periods of rice, with the Fe/Mn oxide fraction having the largest increase and the exchangeable fraction the smallest. The proportion of the residual fraction decreased during the growth periods of rice, while that of the organic matter-bound fraction was relatively stable.

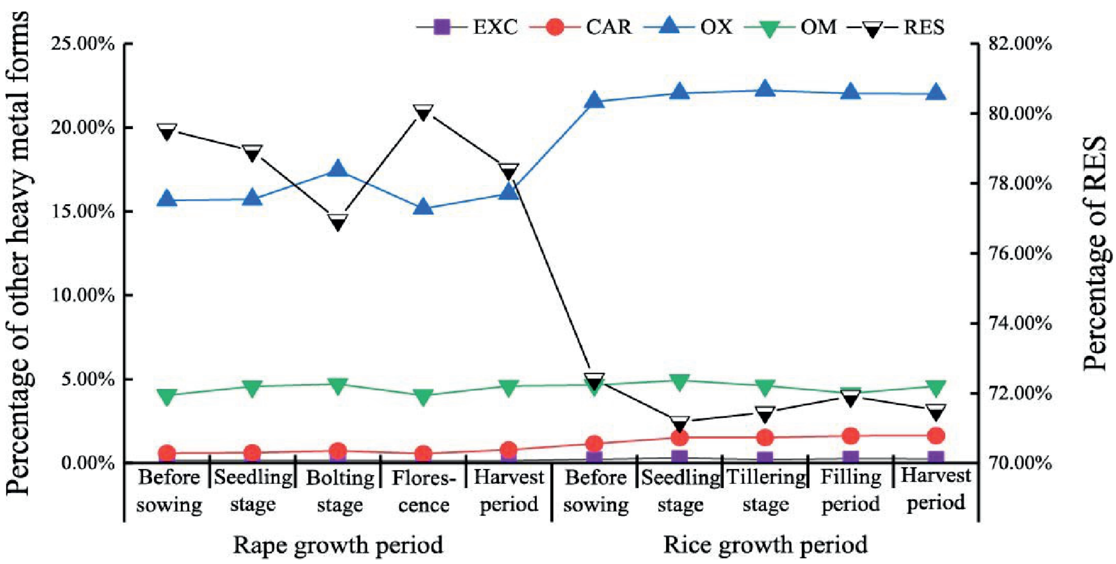

Fig. 1. Variation of Zn speciation in 0-20cm soil during rape rice rotation.

As shown in Fig. 2, among the heavy metal Zn in bottom soil during the rice-rapeseed rotation, the exchangeable and carbonate-bound fractions accounted for low proportions and were relatively stable; the changes in the proportions of the Fe/Mn oxide and residual fractions were completely opposite; the organic matter-bound fraction showed an upward trend during the growth periods of rapeseed, a downward trend during the growth periods of rice, and a slight increase at the rice harvest period. As shown in Fig. 2, the activity of heavy metal Zn in soil tended to increase during the rice-rapeseed rotation. Moreover, the activities of heavy metal Zn in the top and bottom soils were similar at the growth periods of rapeseed but different at the growth periods of rice, with the activity of heavy metal Zn in the top soil greater than in bottom soil.

As shown in Table 3, among the tested heavy metal Zn in the top soil, the exchangeable fraction was significantly positively correlated with the carbonate-bound and Fe/Mn oxide fractions, but significantly negatively correlated with the residual fraction; the carbonate-bound fraction was significantly positively correlated with the Fe/Mn oxide fraction, but significantly negatively correlated with the residual fraction; the Fe/Mn oxide fraction was significantly negatively correlated with the residual fraction, with a correlation coefficient of 0.97. The residual fraction of the heavy metal Zn in the top soil might transform to the other four fractions during the rotation.

As shown in Table 4, during the rice-rapeseed rotation, among the tested heavy metal Zn in bottom soil, the exchangeable fraction was significantly positively correlated with the Fe/Mn oxide and organic matter-bound fractions, but significantly negatively correlated with the residual fraction; the carbonate-bound fraction was significantly negatively correlated with the Fe/Mn oxide, organic matter-bound, and residual fractions; the Fe/Mn oxide fraction was significantly positively correlated with the organic matter-bound fraction, but significantly negatively correlated with the residual fraction; the organic matter-bound and residual fractions were extremely significantly negatively correlated. During the growth periods of both rapeseed and rice, the proportion accounted for by the residual fraction in the heavy metal Zn in bottom soil tended to decrease, and it might transform to the Fe/Mn oxide, organic matter-bound, and carbonate-bound fractions.

Correlation between the soil heavy metal activity and soil physical and chemical properties were analyzed, and the results are shown in Table 5. In the top soil, except that the activity of heavy metal Cu was positively correlated with coarse and medium sand (with a correlation coefficient of 0.64), the other elements were not correlated with the soil mechanical composition; the activities of heavy metals Cr and Pb in soil were negatively correlated with soil pH (with correlation coefficients of -0.69 and -0.66, respectively); the activities of heavy metals Cr, Cd, Pb, and Zn in soil were negatively correlated with soil redox potential (Eh), and in particular, the correlations with Cr and Cd reached an extremely significant level (with correlation coefficients of -0.85 and -0.83, respectively), while the activity of the tested heavy metal Cu in soil was significantly positively correlated with soil Eh (with a correlation coefficient of 0.86); the activities of the tested heavy metals Cr and Pb in soil were significantly positively correlated with soil organic matter (with correlation coefficients of 0.73 and 0.70, respectively); the activity of the tested heavy metal Cu in soil was positively negatively correlated with the organic matter (with a correlation coefficient of -0.70).

In the bottom soil, the activity of heavy metal in soil was not correlated with the mechanical composition; the activity of heavy metal Zn in soil was positively negatively correlated with soil pH (with a correlation coefficient of -0.67); the activities of heavy metals Cr and Pb in soil were extremely significantly negatively

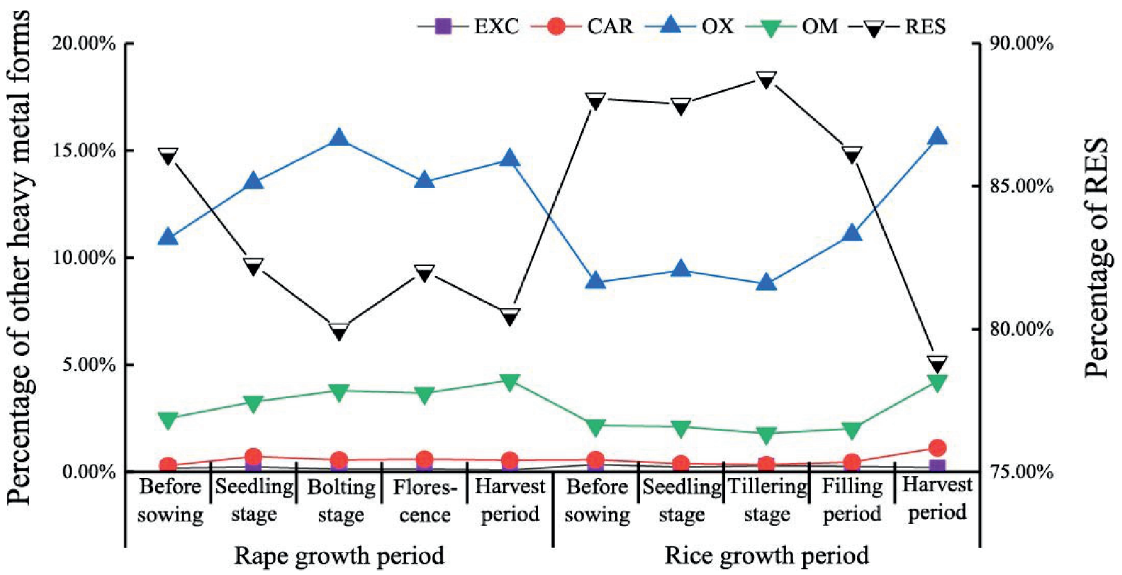

Fig. 2. Variation of Zn speciation in 20-40 cm soil during rape rice rotation.

Table 3. Transformation of heavy Metal Zn forms in surface soil during rice-rape rotation

Zn Speciation	EXC	CAR	OX	OM
CAR	0.92**			
OX	0.92**	0.96**		
OM	0.43	0.36	0.43	
RES	-0.93**	-0.96**	-0.97**	-0.49

Note: EXC is the exchangeable fraction, CAR is carbonate fraction, OX is iron-manganese oxide bound fraction, OM is organic-bound fraction, RES is residual fraction. * Significant at 0.05 level, ** significant at 0.01 level.

Table 4. Transformation of heavy Metal Zn forms in the bottom soil during rice-rape rotation.

Zn Speciation	EXC	CAR	OX	OM
CAR	-0.13			
OX	-0.77**	0.67*		
OM	-0.78**	0.68*	0.95**	
RES	0.74*	-0.72*	-0.99**	-0.97**

Note: EXC is the exchangeable fraction, CAR is carbonate fraction, OX is iron-manganese oxide bound fraction, SOM is organic-bound fraction, RES is residual fraction. * Significant at 0.05 level, ** significant at 0.01 level.

Table 5. Correlation analysis between soil heavy metal activity, morphological distribution and soil physical and chemical properties.

Soil Layer	Mechanical Composition						pH	Eh	SOM
	Clay	FSP	MT	CT	FS	CSM			
0-20 cm	0.15	0.51	0.42	-0.50	-0.18	-0.58	0.09	-0.71*	0.62
20-40 cm	-0.19	0.11	0.15	-0.28	0.27	0.05	-0.67*	-0.28	0.28

Note: FSP is fine sand powder; MT is medium silt; CT is coarse silt; FS is fine sand; CSM is coarse sand and medium sand; SOM is soil organic matter; Eh is electric potential. * Significant at 0.05 level, ** significant at 0.01 level.

Table 6. Stepwise linear regression simulation between soil heavy metal activity and soil physical and chemical properties.

	0-20cm		20-40cm	
	Regression equation	F Test	Regression equation	F Test
Zn	Y = -6.86*10-3 - 1.88*10-5 Eh + 1.09*10 - 3FSP	14.70**	-	-

Note: ** significant at 0.01 level.

correlated with soil Eh (with correlation coefficients of -0.95 and -0.85, respectively), while the activity of heavy metal Cu in soil was extremely significantly positively correlated with soil Eh (with a correlation coefficient of 0.90); the activity of heavy metal Cu in soil was significantly positively correlated with soil organic matter (with a correlation coefficient of 0.75).

Discussion and Conclusions

This study showed that the whole amount of heavy metal Zn ranged from 74.01 mg·kg^{-1} to 82.16 mg·kg^{-1} during the rice-oilseed rape rotation, which was higher in this study area compared to other provinces in China, such as Guangxi (43 mg·kg^{-1}), Guangdong (60 mg·kg^{-1}), and Henan (63 mg·kg^{-1}), this mainly originated from the endogenous type of parent material and the combined effect of natural soil formation process and strong mineral activities in the karstic lime area, but it was lower than the Zn concentration in Shanghai (103 mg·kg^{-1}), which was due to the fact that Shanghai is located in the Yangtze Delta area, the terrain is flat and the local area of lakes and marshes depressions are densely populated, which is a geological environment favorable to the deposition of Zn, and makes the local area of Shanghai with a good concentration of Zn [15].

At the same time, in the experiment, we also observed that the total amount of heavy metal Zn in the surface soil was higher than that in the bottom soil, showing a certain surface aggregation. Some studies have shown that soil temperature has a significant effect on the adsorption of soil Zn, and the increase in temperature provides enough energy for the soil to adsorb Zn, thus increasing the adsorption capacity of the soil, but the increase decreases gradually with the increase in soil depth. The reason for this phenomenon may also be related to the soil mechanical composition and SOM content, because the soil mechanical composition will have an impact on the distribution of SOM, and SOM will be complexed with Zn, which will change the adsorption performance of the soil on Zn, so it led to a higher concentration of Zn in the surface layer, and at the same time the SOM content is also higher, but with the depth of the depth of the increase, the two at the same time decreased [16].

In the surface soil during rice-oilseed rape rotation, the total amount of heavy metal Zn ended up being similar to that of oilseed rape and rice at the pre-sowing and harvesting periods, regardless of the large fluctuations in the total amount of heavy metal Zn during the different growing periods of rice and oilseed rape. This suggests that crop effects on total soil heavy metal Zn occur mainly during the sowing to harvest period. After harvest, the total amount of heavy metal Zn in the soil remained at its original level. This phenomenon also existed during the growing period of oilseed rape in the subsoil. Some studies have shown that oilseed rape has a very strong heavy metal enrichment ability [17] and can be used as a remediation plant in heavy metal polluted areas, while rice also has a strong bioconcentration of Zn [18], so the effect of crops on Zn mainly occurs during the sowing to harvesting period.

In this study, the residual fraction of heavy metal Zn accounted for the largest proportion in the 0-40 cm layer of soil, which was consistent with findings of [19]. The author of this article was more inclined to the findings of Zhang et al., possibly because of the geographical location of the trial sites. This study and that of Zhang et al et al. were both located in the suburbs, but the study of other researchers [20] was located along the highway. Although they were all affected by human activities to a large extent, the different ways and methods of influence might lead to different findings. In the 0-40 cm layer of soil, the exchangeable fraction of heavy metal Zn accounted for a small proportion, which is consistent with the findings of Zhang et al et al [21]. During the growth periods of rapeseed, the residual fraction changed similarly in the 0-20 cm and 20-40 cm layers of soil. However, during the growth periods of rice, the residual fraction changed almost oppositely in the 0-20 cm and 20-40 cm layers of soil, which might be related to the flooding. During the rice-rapeseed rotation, the Fe/Mn oxide fraction of heavy metal Zn in the top soil accounted for a large proportion, and the trend of its change was consistent with the activity of heavy metal Zn.

In this study, among the tested heavy metal Zn in the top soil, the exchangeable fraction was significantly positively correlated with the carbonate-bound and Fe/Mn oxide fractions, and significantly negatively correlated with the residual fraction. However, among the heavy metal Zn in bottom soil, the exchangeable fraction was significantly positively correlated with the Fe/Mn oxide, organic matter-bound, and residual fractions. This might be due to the fact that Eh in the bottom soil is much higher than that in the top soil, making the residual fraction of heavy metal Zn in the bottom soil increase substantially [22]. From the soil surface to 0-40 cm below vertically, it was found that the residual fraction of heavy metal Zn in soil was significantly negatively correlated with the carbonate-bound, Fe/Mn oxide, and organic matter-bound fractions. Particularly, the residual fraction was significantly negatively correlated with the corresponding organic matter-bound fraction in the bottom soil.

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Conflict of Interest

The authors declare no conflict of interest.

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